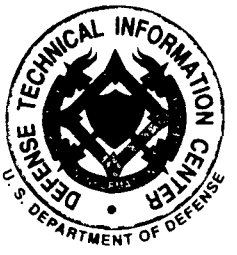

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LIQUID VISCOSITY AND DENSITY MEASUREMENT WITH FLEXURAL-PLATE-WAVE SENSORS

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Abstract: Micromachined flexural-plate-wave (FPW) sensors can detect subtle variations in the density and viscosity of liquids. Liquid density causes a mass-loading effect that lowers the sensor operating frequency. In viscous liquids, the attenuation coefficient of the acoustic waves is proportional to the square-root of the viscosity.

We discuss recent results that explore the density-sensing precision and the viscosity-sensing range of FPW sensors. A micromachined FPW sensor (wavelength 100 μm, plate thickness 6.0 μm, operating frequency in water 5.7 MHz) detected the density of a variety of solvents and aqueous salt solutions with an error less than ±0.003 g/cm^3. Most of this error was likely due to the uncertainty with which we know the true density and sound velocity of each sample.

The same sensor was used to measure the viscosity of aqueous polymer solutions (poly(ethylene glycol) and hydroxyethyl cellulose). The sensor accurately measured the viscosity of solutions with molecular weights up to around 10,000. Polymer solutions with MW > 20,000 appeared water-like to the sensor (i.e., there was little or no attenuation of the acoustic waves).The decrease in apparent viscosity measured with the FPW sensor occurs for polymers with relaxation frequencies greater than or equal to the FPW frequency.

Key Words: Densitometer; density; flexural; oil; sensor; ultrasonic; viscometer; viscosity

Introduction: The density of liquids has traditionally been measured by either glass hydrometers or vibrating mechanical elements; viscosity has been measured with expensive laboratory instruments such as cone-and-plate viscometers, or simple glass apparatus such as capillary viscometers. All require large sample volumes, and non are amenable to continuous, on-line measurement.

Previously we have reported a silicon-based microsensor that can continuously measure the density and viscosity of tiny quantities of liquids [1][2][3]. The microsensor, shown in Fig. 1, utilizes ultrasonic flexural-plate waves (or Lamb waves), one of a family of acoustic modes that have been exploited for measuring liquid properties. Others in the family are shear-mode bulk acoustic waves [4][5][6], shear-horizontal acoustic plate waves [7], Love and Bleustein-Gulyaev surface waves [8], and torsional waves [9]. The flexural plate wave (FPW) differs from these other modes in that the wave

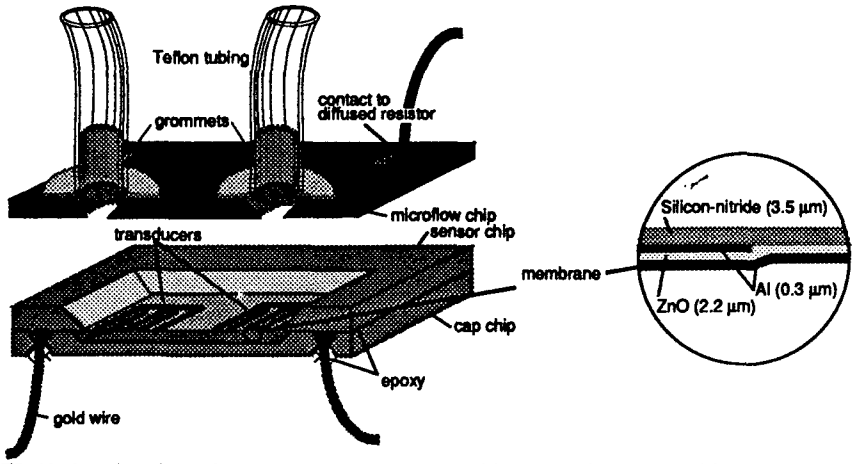

FIGURE 1. FPW density/viscosity sensor. The sensor chip, containing the thin plate or membrane, is attached to a cap chip which protects the metalized side of the sensor. A third chip forms an enclosure that contains liquids on the sensor, and has a diffused resistor for measuring liquid temperature.

motion has a component of motion that is normal to the sensor surface, as depicted in Fig. 2. The interaction between FPWs and an overlying liquid is primarily due to this normal motion rather than a shear interaction at the sensor surface. This makes the FPW sensor much more responsive to liquid density, even in inviscid liquids.

The FPW sensor has been used to measure the viscosity of dimethyl sulfoxide solutions at near-freezing temperatures [10] and to follow the density change associated with the fermentation of glucose by yeast [11] and diffusion in gels [12]. Here we describe the FPW sensor response to viscous fluids and quantify the accuracy with which prototype FPW sensors can measure the density of liquids. We also investigate the relation between the FPW-measured viscosity of polymer solutions and the molecular weight of the polymer.

Theory of operation: Figures 1 and 2 shows the key element of the FPW sensor: a thin plate that supports a traveling flexural wave. Interdigital transducers (IDTs) on either end of the membrane are used to excite and detect flexural waves, whose properties—such as velocity and attenuation—are affected by the measurand of interest. We use the IDTs to measure (1) center or synchronous frequency, which is proportional to wave velocity, and (2) ratio of output to input voltage, which is dependent on wave attenuation.

A simple and accurate approach for modeling sensor response is to solve a thin-plate differential equation of motion for the wave-propagation characteristics. This method yields simple, analytical expressions for the flexural-wave phase velocity in multilayered plates subjected to in-plane tension. It can be extended to include the effects of a viscoelastic medium contacting the plate, yielding simple expressions for the phase velocity and attenuation coefficient when the medium is a viscous or inviscid liquid. Here we summarize a thin-plate solution derived previously; for the full derivation see References [3] and [13].

1. **Free plate:** Using the coordinate system shown in Fig. 2, we derive an approximate one-dimensional equation of motion by balancing the forces acting on a plate segment. Within thin plates, the dominant forces arise from the flexural rigidity D and tension T in the plate, and from

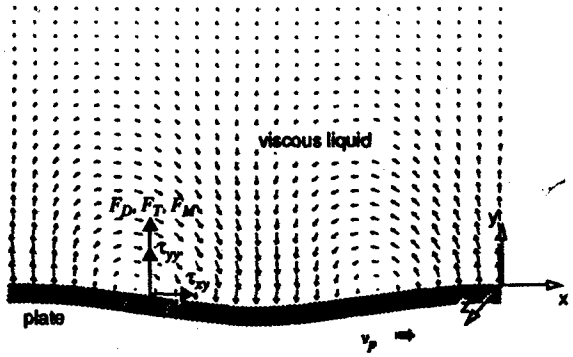

FIGURE 2. Forces acting on a plate segment are balanced to derive an equation of motion for the flexural wave, from which the phase velocity and attenuation coefficient can be calculated (see text). Stresses τ_{yy} and τ_{xy} induced in the viscous medium by the plate motion impede the transverse and rotational motion of the segment, respectively. The arrows represent the displacement or velocity field.

the resistance of the segment mass M to acceleration. We designate these forces (per unit plate area) F_D, F_T and F_M, respectively. Expressions for these terms are found with plate theory, assuming a harmonic traveling-wave with normal displacement

$$u_y = |U|e^{j(\omega t - kx)} = |U|e^{-\alpha x}e^{j(\omega t - \beta x)}. \quad (1)$$

Here, ω is the angular frequency, k is the complex propagation constant, and α and β are the attenuation coefficient and wave number, respectively, both of which are real quantities. Wave number is related to the wavelength λ of the flexural wave by

$$\beta = 2\pi/\lambda. \quad (2)$$

In typical operation, wavelength is fixed by the transducer period P ($\lambda \equiv P$) and thus β is constant.

For a freely traveling wave, the forces acting on the plate must sum to zero, which yields:

$$v_p = \frac{\omega}{\beta} \cong \left(\frac{T + \beta^2 D}{M}\right)^{1/2}, \text{ and } \alpha = 0, \quad (3)$$

where v_p is the phase velocity. For a homogeneous plate of thickness d, Young's modulus E, Poisson's ratio ν, in-plane stress σ and mass density ρ, $D = Ed^3/12\left(1-\nu^2\right)$, $T = \sigma d$ and $M = \rho d$.

2. Inviscid liquids: When an inviscid liquid contacts the plate, it impedes the normal plate motion, adding an additional force term to that of the free plate. Summing the plate forces to zero in this case yields several possible modes with different attenuation coefficients. For density sensing, we are interested in the lossless mode, also called a "Scholte" mode, for which $\alpha = 0$:

$$v_p = \frac{\omega}{\beta} \cong \left(\frac{T + \beta^2 D}{M + \rho\delta}\right)^{1/2}, \text{ and } \alpha = 0. \quad (4)$$

Here,

$$\delta = \left[\beta\sqrt{1 - (v_p/c)^2}\right]^{-1} = \lambda/\left[2\pi\sqrt{1 - (v_p/c)^2}\right] \quad (5)$$

is a *real* number representing the decay depth of the *evanescent* displacement field in the fluid (plotted in Fig. 2). Thus, an inviscid liquid causes a mass loading $\rho\delta$ that lowers the phase velocity and frequency. For typical plate-wave structures (plate thickness 2-5 μm and transducer period P=100 μm) we find that $\delta \equiv \lambda/2\pi \equiv P/2\pi$, and that $\rho\delta$ is of the same order as the plate mass per unit area, M. This results in very large frequency and velocity shifts when liquids are introduced to the plate surface, and is the reason for the extraordinarily high density sensitivity of FPW sensors.

3. **Viscous liquids:** When the liquid has a finite viscosity, the retrograde elliptical particle motion of the plate induces both reactive normal stress (τ_{yy}) and shear stress (τ_{xy}) in the liquid that act on the plate surface. The shear stress τ_{xy} produces a moment that impedes the plate segment's rotary acceleration. Using linear elastodynamics and assuming that: (i) the particle displacement across the interface at $y = 0$ is continuous (the so-called "no-slip" condition); and (ii) the position of the neutral plane of the plate is unaffected by the presence of the viscoelastic medium, the force-balance equation yields:

$$v_p = \frac{\omega}{\beta} \cong \left(\frac{T + \beta^2 D}{M + \rho\delta + M_\eta} \right)^{1/2}, \text{ and } \alpha \cong \left(\frac{\omega}{2}\right)^{3/2} \frac{\delta^2 \beta \sqrt{\eta\rho}}{T + 2\beta^2 D + \omega^2 \rho\delta/\beta^2}, \quad (6)$$

where η is the shear viscosity and $M_\eta = (\beta\delta)^2\sqrt{\eta\rho/2\omega}$ is the effective mass contributed by the liquid viscosity. Assumption (ii) is valid if the magnitude of the viscous shear modulus $\omega\eta$ is small compared to the elastic moduli of the plate materials.

The fluid viscosity therefore adds additional mass loading, and causes wave attenuation. Note that, as we would expect, Eq. (6) reduces to the case of an inviscid fluid (Eq. (4)) when η=0, and to that of a free plate (Eq. (3)) when $\rho = \eta = 0$.

In practice, only ρ and η are unknowns in Eq. (6), and we have to take two measurements—of ω and α—to determine both. We operate the sensor at a constant wavelength $\lambda \equiv P$, where P is the transducer period. This constrains $\beta = 2\pi/\lambda$ to a known constant. The plate parameters D, T and M are measured with a simple calibration procedure, discussed later. We also assume that $\sqrt{1 - (v_p/c)^2} \cong 1$, since typically $v_p \ll c$.

Sensor Fabrication: Each FPW delay line consisted of a composite thin-film membrane (Fig. 1 inset). Silicon wafers were coated with 3.5-μm-thick LPCVD low-stress silicon nitride and etched in KOH, leaving membranes and 3 mm by 8 mm in 500-μm-deep wells. Sputtered aluminum was patterned into interdigital transducers, having period P=100 μm, which were then coated with RF-magnetron-sputtered zinc oxide (2.2 μm thick) and a 0.3-μm-thick aluminum ground plane. This sensor had a synchronous frequency of 7.6 MHz when operated in air, and 5.7 MHz when in contact with deionized water on one side.

Since FPW sensors are equally sensitive to the environment on both sides of the membrane, a second encapsulation chip was fabricated to hermetically seal one side of the membrane and to protect the aluminum and zinc oxide from corrosion. The sensor and cap chips were bonded to each other with epoxy

(Epo-tek 302) that was spin-coated onto the cap wafer. Electrical contact was made to the sensor with 100 μm gold wires indium-soldered to the bond pads and then coated with epoxy.

A third chip encased the well-side of the sensor and had two etched holes to which grommets were epoxied. Liquids were introduced to the sensor via Teflon tubing fitted over the grommets. This chip also contained a diffused resistor whose resistance was used to monitor the temperature of the liquid in contact with the sensor. The composite chip was attached to an aluminum carrier that was bolted to a temperature controlled copper block (set point 25 °C).

Density Measurement: The change in the frequency of an FPW delay-line oscillator was recorded as the sensor was exposed to a number of organic solvents. The data were corrected for temperature using the readings from the temperature-measuring resistor, and then fitted to Eq. (4) to extract the parameters M and $B = T + \beta^2 D$. A comparison of the FPW measurements and the published values of the liquid densities appears in Fig. 3A. The discrepancies are plotted in Fig. 3B. Notice that the scatter in the measurements of density of each solvent is much less than the sample-to-sample error.

In order to have this level of agreement we need to know the speed of sound c in the fluid (which enters into the expression for δ). We can circumvent this problem in at least two ways: (1) design the sensor with lower phase velocity so that it is less sensitive to the speed of sound in the liquid, and then assume a constant c; (2) measure the sound velocity with two FPW sensors operating at different wavelengths. For many applications, assuming a constant speed of sound should be adequate. For example, if all the liquids being tested have similar sound speeds or if the sound speed is correlated with liquid densities, then the effect of sound speed can be ignored. This principle has been demonstrated by measuring the density of salt solutions. The densities of the solutions, 0 to 100 g/l NaCl in 25 g/l increments, were calculated assuming a constant speed of sound of 1497 m/s. The error (Fig. 3B) was again only about ± 0.2%.

There are two sources of error in both these examples. There is a small amount (less than 0.001 g/cm^3) of scatter between repeated measurements of the same sample. This seems to be related to the introduction and removal of liquids from the sensor surface. There is also sample-to-sample error that may be caused by the difference between the actual densities of the samples and the literature values that we used in our calculations. Both errors are absent when FPW sensors are used to measure the change in the density of a solution that is in continual contact with the sensor, as would be the case in continuous, in-situ density monitoring.

We have seen this improvement, effectively, when we monitored sodium chloride diffusion through a polyacrylamide gel. The gel, made with deionized water, was polymerized on the sensor surface. The deionized water covering the gel was replaced with a 0.1 M NaCl solution. The sensor frequency dropped as salt diffused through the gel toward the sensor surface. Experiments like this have been used to measure the diffusion coefficients of a number of gels and solutes [12]. The noise level in this experiment corresponds to density perturbations of only 0.0001 g/cm^3.

Viscosity Measurement: We measured the FPW sensor response to different viscous liquids. Frequency of maximum transmission (ω) and corresponding insertion loss were determined by driving one IDT pair with a 25-cycle RF tone burst while sweeping the RF frequency and measuring the voltage of the burst received on the second IDT with a digital oscilloscope. Insertion loss was determined

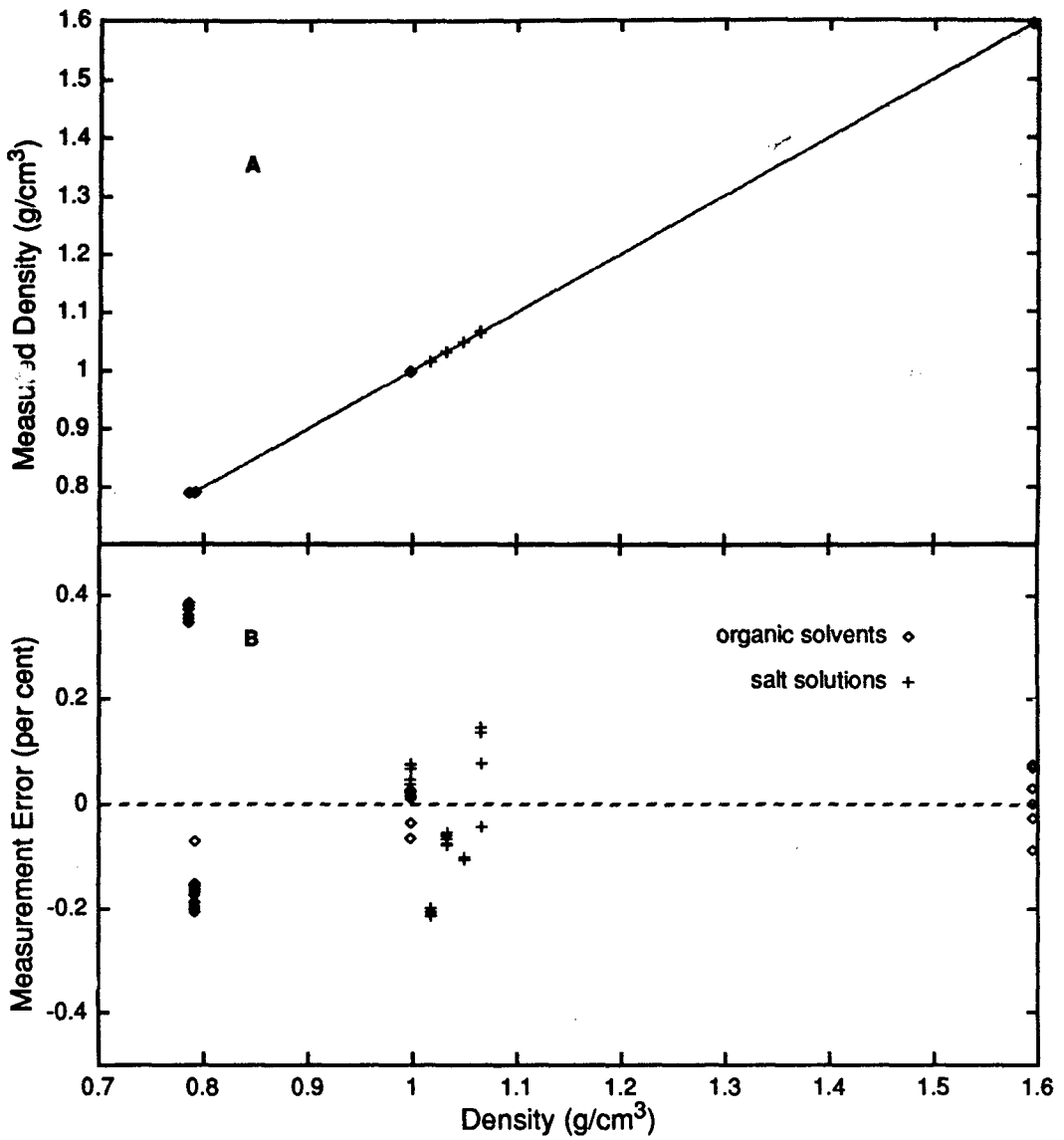

FIGURE 3. FPW-measured density versus literature values for methanol, acetone, deionized water, carbon tetrachloride, and aqueous salt solutions (A). The difference between the measured and literature density values are plotted in (B).

from the IDT RF voltage magnitudes by IL=20 log (v_{out}/v_{in}). Attenuation coefficient was calculated from the change of insertion loss $\Delta IL=20\log(e^{-\alpha L})=-8.686\alpha L$, caused by introduction of a viscous liquid on the sensor, where L=5 mm is the distance between IDTs.

The low-frequency, or low-shear-rate viscosities of all solutions were measured independently with a capillary viscometer. Glycerol-water solutions were used for reference, because of their frequency-independent (Newtonian) viscosity characteristics. The FPW-measured viscosity, determined from measured ω and α and Eq. (6), is plotted against the capillary-viscometer-measured viscosity in Fig. 4.

The data from solutions of poly(ethylene glycol), having average molecular weights 3350 and 15,000, and glycerol-water fall on the same line. The FPW-measured viscosity of higher-molecular-weight polymers is lower than the capillary-viscometer-measured viscosity. This effect is most pronounced with the 98,000 molecular-weight hydroxyethyl cellulose which shows near-water apparent viscosities. We have seen similar results for the FPW-measured viscosity of salmon-sperm DNA solutions.

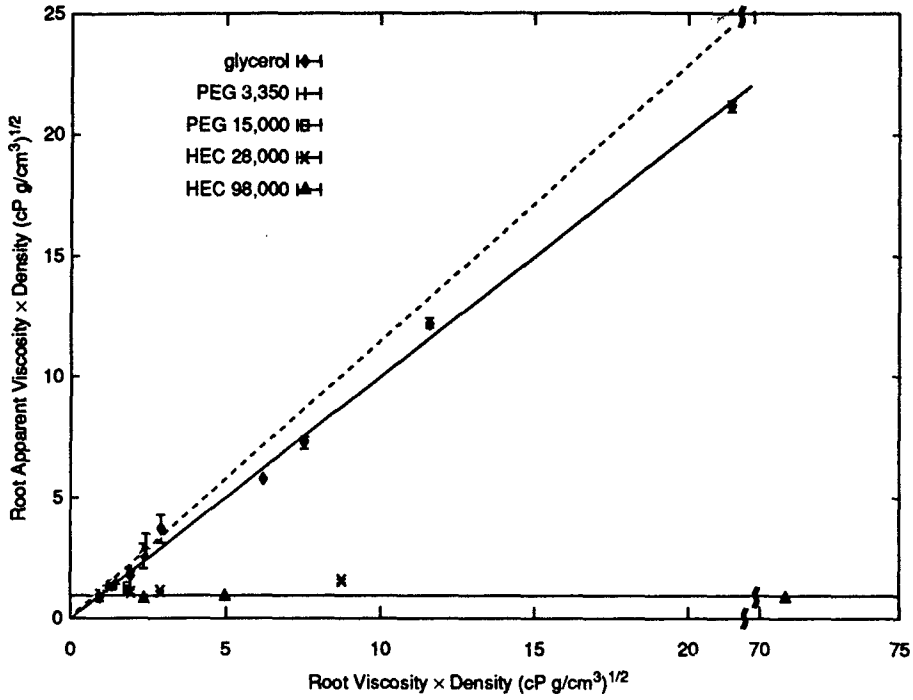

FIGURE 4. Plot of the viscosity-density product of a number of aqueous solutions of the polymers poly(ethylene glycol) (PEG) and hydroxyethyl cellulose (HEC). The response of the FPW sensor (vertical axis) is calibrated using glycerol solutions and is compared to measurements made with a capillary viscometer (horizontal scale). Error bars represent the maximum and minimum values observed. The dotted line represents the attenuation predicted by an approximation of Eq. (6) that assumes ω and δ are constant. This approximation is only valid at lower viscosities; at higher viscosities, ω decreases as viscosity increases, and the line over predicts the FPW-measured viscosity.

Discussion: Our results have demonstrated the sensitivity with which the FPW sensor responds to both density and viscosity of liquids. For low-viscosity liquids (like water) the effect of viscosity on the oscillation frequency is small. For the sensor used in this experiment, a viscosity increase of 0.3 cP would only lower the frequency by 1.3 kHz – the same as a density change of 0.001 g/cm^3. The same viscosity change would increase the transmission loss by 2.4 dB. Higher viscosity liquids increase the FPW attenuation and lower the phase velocity. By observing both quantities, it is possible to calculate both the viscosity and the density of the liquid.

The behavior of the FPW sensor when exposed to high molecular weight solutions is similar to that observed by Reed et al. with a shear-thickness mode quartz resonator [5]. They used a variation on the Maxwell model for the complex shear modulus of a viscoelastic medium to account for the lower than expected viscosity of high molecular weight polymers:

$$\eta = \eta_0 \frac{j\omega}{(1 + j\omega\tau)^{\gamma}}, \tag{7}$$

where τ is a viscoelastic relaxation time characteristic of the polymer and γ represents the distribution of relaxation times. We have shown previously [12] that the FPW sensor is relatively insensitive to the real shear modulus of an overlying solid (such as a gel) so long as the shear modulus is less than 10^6 Pascals. Assuming the attenuation we observed is due entirely to the imaginary part of the shear modulus and $\gamma = 1$ (a Maxwellian fluid), we have calculated a viscoelastic relaxation time of 830 ns for the 24,000 molecular-weight HEC. This is consistent with the relaxation time of solutions of polymers with similar molecular weights [14].

While this aspect of FPW behavior is similar to that of other acoustic sensors, the mechanism of the FPW interaction with liquids is unique. Shear stress at the interface of the sensor and the liquid (τ_{xy} in Fig. 2) has relatively little effect on the velocity or attenuation of the plate waves. This could have the benefit of making the FPW sensor less affected by surface properties (such as roughness or hydrophobicity) than shear-mode acoustic sensors.

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